GEL COUNTING OF ^{14}C AND ^{3}H*

T. M. KRISHNAMOORTHY AND S. D. SOMAN

Health Physics Division, Bhabha Atomic Research Centre, Bombay 400 085

ABSTRACT

Aluminum stearate gel in toluene base scintillator is employed for the measurement of varied amounts of carbon–14 labelled $BaCO_3$ and tritiated lysine and the counting efficiencies are compared with the solution counting. $BaCO_3$ particulates in the ranges of $74 < X < 105$ μ. dia. and $105 < X < 416$ μ dia. do not affect the gel counting efficiency. Sample weights upto 100 mgm do not cause any significant change in the gel counting efficiency while further increase in weight definitely lowers the efficiency. A comparison is made of the direct counting of carbon-14 labelled $BaCO_3$ particulates with that of the gel suspension counting. Gel counting efficiency for tritium is found to be of the order of 1% of the solution counting. For carbon-14 this is about $92 \pm 4\%$ of the solution counting in the weight range upto 100 mg $BaCO_3$.

INTRODUCTION

LIQUID scintillation counting technique is the most common method in the recent years for the measurement of soft beta activity. In fact, any radionuclide that can be incorporated into or suspended in a scintillator solution, can be counted with a liquid scintillator system. The scope of liquid scintillation technique is enhanced with the advent of numerous heterogeneous systems[1-8] wherein the problems of solubility or quenching are significantly eliminated.

In the present investigations, a suspension technique using aluminum stearete gel in toluene, PPO and POPOP scintillator system is tried for the measurement of ^{14}C and ^{3}H in solid samples and the counting efficiencies relative to solution counting are obtained. For this purpose, ^{14}C labelled $BaCO_3$ and tritiated lysine are used. A comparison is made of the direct counting of $Ba^{14}CO_3$ in toluene scintillator with that of the gel suspension technique.

MATERIALS AND METHODS

Gel preparation.—Toluene scintillator was prepared with 6·5 gm PPO and 0·13 gm POPOP in one litre distilled toluene. To 10 ml of this scintillator, 1·5 gm aluminum stearate was added, mixed thorougly and heated in a water-bath at 80° C for 5 minutes[1]. The gel formed was uniform and even after stirring with the suspended particulates, attains uniformity immediately without air bubbles. Similar gel prepared with dioxan scintillator (6·5 gm PPO, 0·13 gm POPOP, 100 gm naphthalene in one litre distilled dioxan) showed less uniformity and was unstable after shaking; hence the former was used. The active material was incorporated into the toluene scintillator solution prior to gelling and this resulted in a fairly uniform distribution of the sample in the gel.

^{14}C labelled $BaCO_3$.—^{14}C was obtained from the Isotope Division, BARC, in the form of Na_2CO_3 (8 uci/110 μgm Na_2CO_3). A stock solution was prepared from this, containing $4 \cdot 4 \times 10^4$ dpm/ml. A definite amount of this activity was added to known amounts of inactive Na_2CO_3 (10 mg/ml) and mixed thoroughly. Excess $BaCl_2$ solution was added to precipitate $BaCO_3$. The precipitate was coagulated by heating in water-bath at 90° C for 5 minutes, centrifuged and washed with distilled water thoroughly to remove the excess $BaCl_2$ reagent. The precipitate was then dried under infrared lamp, powdered, sieved and used for the experiments.

Gel counting of $Ba^{14}CO_3$.—A bulk amount of labelled $Ba^{14}CO_3$ was prepared using known amounts of inactive sodium carbonate and ^{14}C activity. The $BaCO_3$ precipitate was dried, powdered and sieved through Greenings Test meshes 25 and 36 (Middlesex U.K.), to get the particle sizes in the range of $416 < X < 675$ μ dia. Varied amounts (20–370) mgm of these particulates were suspended in toluene-aluminum stearate scintillator gel and counted to study the effect of weight in gel counting efficiency (Fig. 1). An aliquot of the added ^{14}C activity was measured using dioxan scintillator for comparison (solution counting).

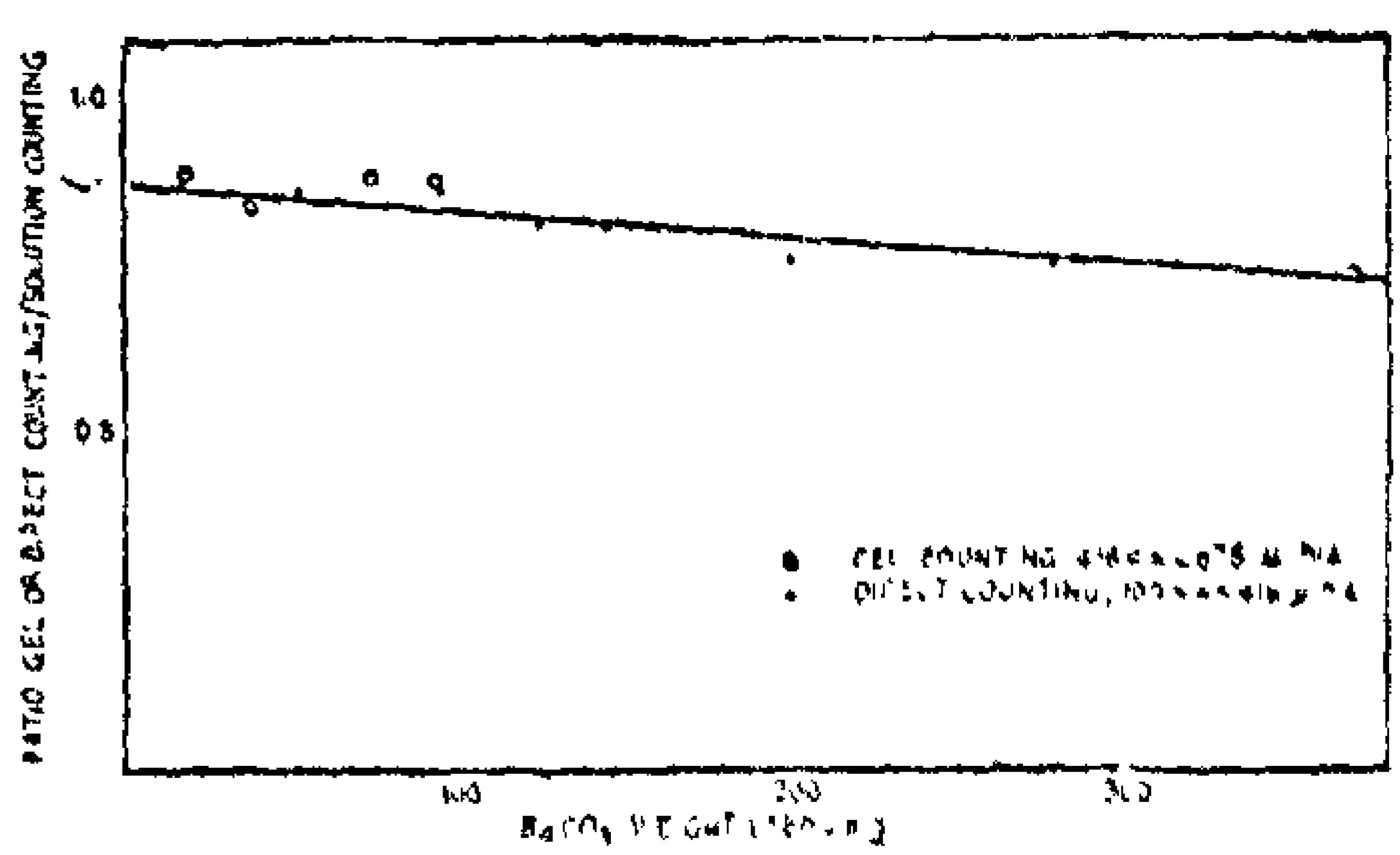

FIG. 1. Heterogeneous counting efficiency *vs.* weight.

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In order to study the effect of change in the specific activity of $Ba^{14}CO_3$ in gel counting efficiency, the following experiments were conducted: A set of 5 ml Na_2CO_3 solutions, containing 50 mgm Na_2CO_3 each ($\sim$ 90 mgm $BaCO_3$), were spiked with different amounts of ^{14}C activity and labelled $BaCO_3$ was prepared as described earlier. The $Ba^{14}CO_3$ particulates thus prepared were sieved to obtain particle size of $74 < \times < 105$ μ dia. and $105 < \times < 416$ μ dia. The particles were suspended in the stearate gel, counted and compared with the solution counting of the added ^{14}C activity. A similar experiment was performed with the particle size of $416 < \times < 675$ μ dia. using 370 mgm of $BaCO_3$ (Fig. 2).

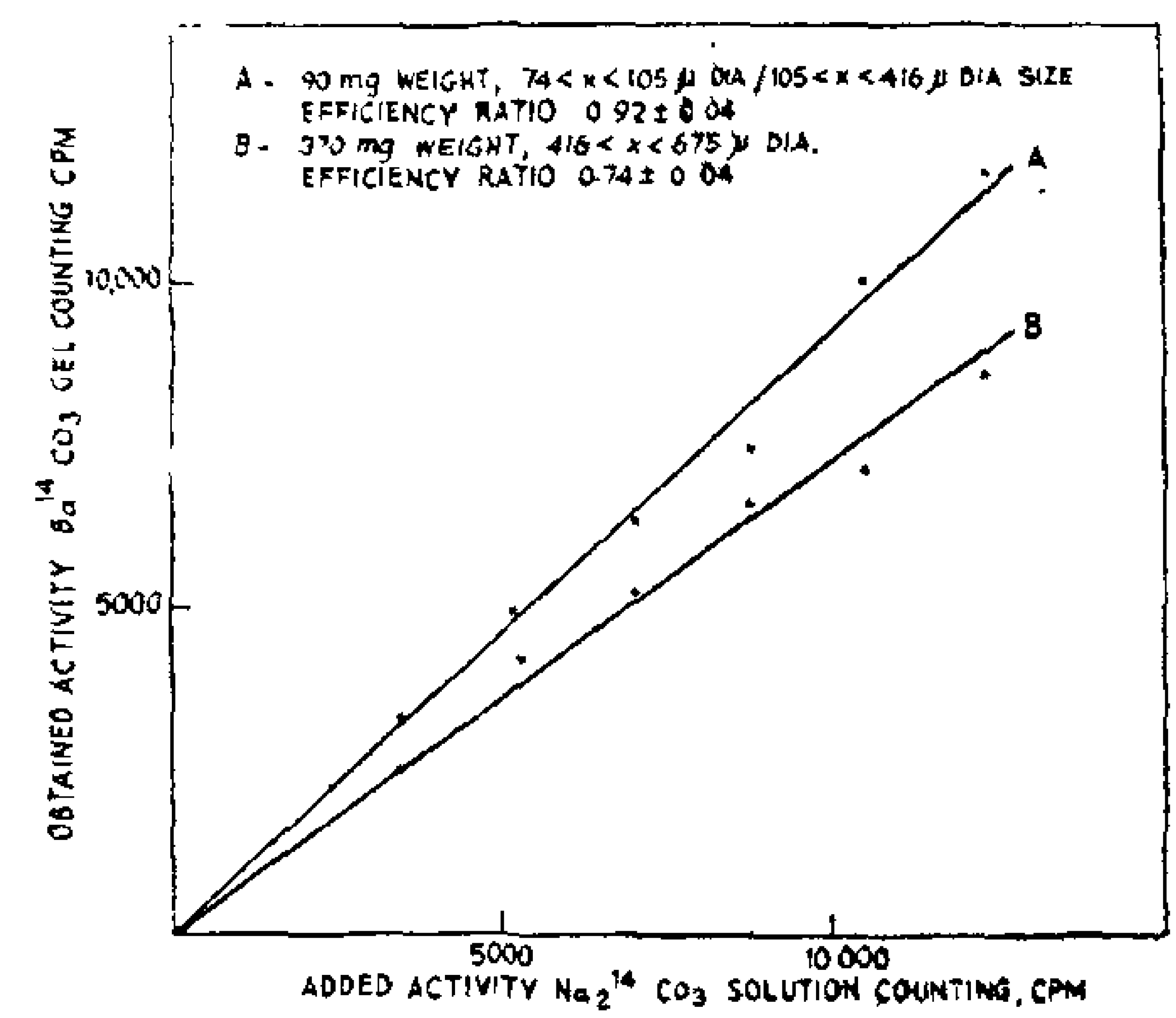

FIG. 2. Gel Counting Efficiency for varied specific activity of $Ba^{14}CO_3$.

Direct counting of $Ba^{14}CO_3$.—A study was made to determine the direct counting effeciency for $Ba^{14}CO_3$ particulate suspensions in toluene scintillator system. Varied amounts of $Ba^{14}CO_3$, with particle sizes of the order of $105 < \times < 416$ μ dia. were used. The counting efficiency was computed relative to the counts obtained for aqueous ^{14}C activity in dioxan scintillator (Fig. 1). Table I gives the direct counting efficiency for different amounts of labelled $BaCO_3$ of varying particle sizes.

TABLE I

Direct counting of $Ba^{14}CO_3$ in toluene scintillator

$BaCO_3$ mg	Ratio of $BaCO_3$	Solid counting w.r.t.		Solution counting
	74< ×<105	particle size in μ dia. 105< ×<416	416< ×<675	675< ×<1000
20	0·93	..	0·85	0·78
40	0·93	0·87	0·85	0·80
100	..	0·88	0·86	0·81
150	..	0·82	..	..
200	..	0·77	..	..
280	..	0·76	..	..

Gel counting of ^{3}H.—Tritiated toluene obtained from the Isotope Division, BARC, was diluted with toluene to give 1100 dpm per ml. 10 ml of toluene scintillator was spiked with 1–5 ml of tritiated toluene. These were initially liquid counted. Different amounts of aluminum stearate (1–2 g) were added to these solutions, and gels prepared, and counted to study the effect of stearate gel on the count rate. The counting losses obtained might be either due to the absorption of the soft betas in the gel or due to the quenching of aluminum stearate. Quenching due to aluminum stearate was studied using 10 ml spiked scintillator and varied amounts of aluminum stearate (0·3–2·1 g) (Fig. 3).

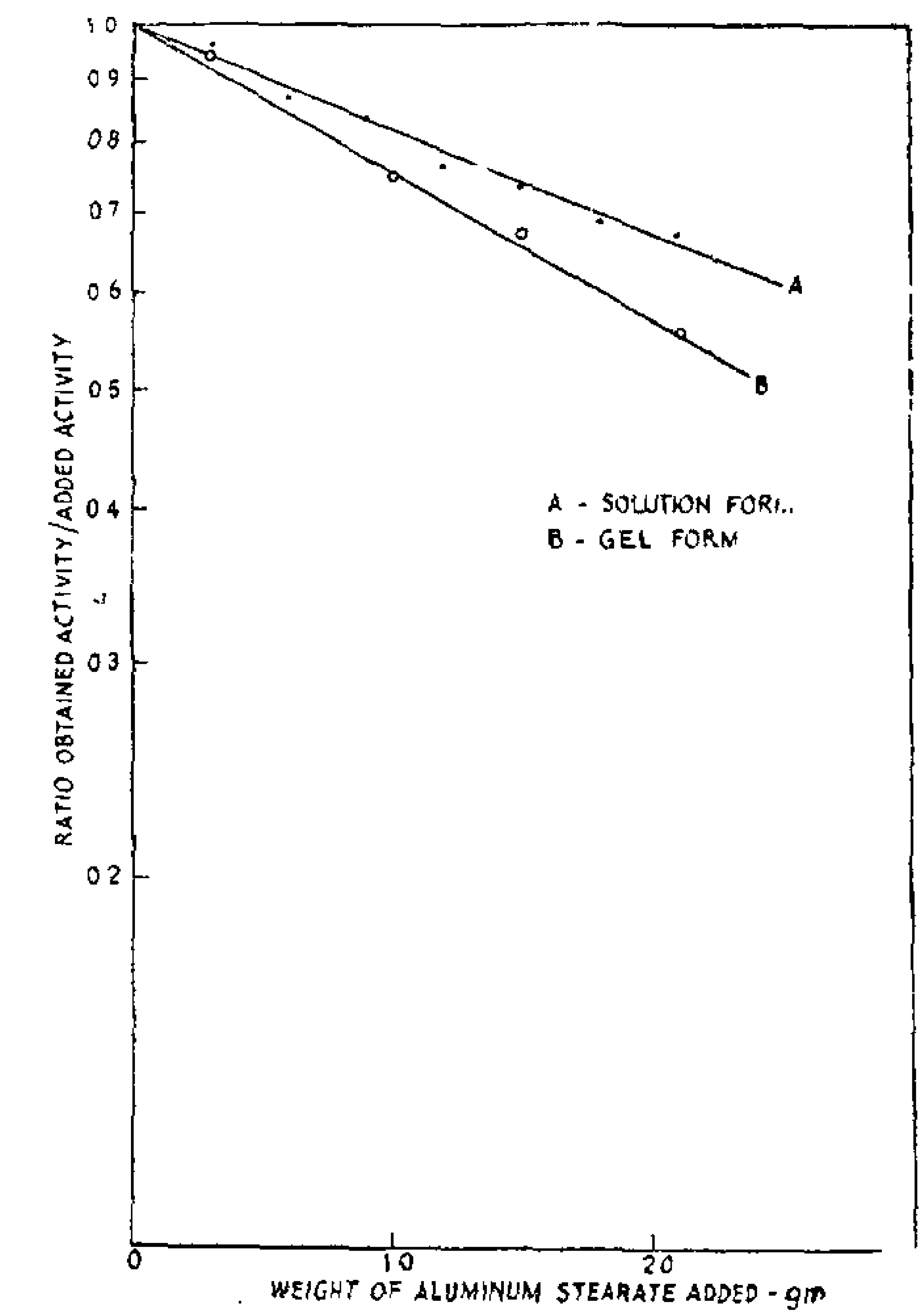

FIG. 3. Quenching due to Aluminum stearate in solution and gel form for tritium.

In order to study the effect of solid suspension of tritiated samples in the gel, 1 mc tritiated lysine with a specific activity of 1200 mc per millimole was diluted and a known volume was mixed with 200 mg of inactive lysine. The labelled lysine was recrystallised from ethanol medium, vacuum dried and weighed. A known portion of the solid was dissolved in water to give 1 mg per ml concentration. This was counted in dioxan scintillator to give the added amino acid activity (solution counting). Different amounts of labelled lysine (12–66 mgm) were suspended in toluene-stearate gel and counted. The ratio between the two would indicate the gel counting efficiency for tritium compared to the solution counting (Table II).

TABLE II

Gel counting efficiency of tritiated lysine relative to the solution counting

Weight of amino-acid added mg	Activity of the amino-acid		Ratio of gel count to solution count
	Solution count cpm	Gel count cpm	
12·1	11080	141	0·0127
25·0	22880	250	0·0109
39·6	36230	391	0·0108
66·5	60850	640	0·0105

RESULTS AND DISCUSSION

The weight *vs* efficiency relation by the gel counting or direct counting technique is shown in Fig. 1. Increased sample weights decrease counting efficiency. Even as large samples as 369 mg could be counted with an efficiency of 0·74 (ratio) of that of solution counting. The direct counting follows the same trend. Nathan *et al.*[9] have reported a loss of less than 25% for five fold weight increase using thixcin-toluene system in 200–1000 mgm range.

Figure 2 shows the linearity of the gel counting efficiency for the varied specific activity of $Ba^{14}CO_3$. For particle sizes of $74 < \times < 105\ \mu$ dia. and $105 < \times < 416\ \mu$ dia., the efficiency is found to be 0·92 when 90 mgm sample used. The same has decreased to 0·74 for the particles in the range of $416 < \times < 675\ \mu$ dia. for a sample weight of 369 mg This may either be due to the increased sample weight or due to the increased particle size. However, from the present studies it can be concluded that particles upto the sizes of $105 < \times < 416\ \mu$ dia. will not significantly affect the counting efficiency. White and Helf[2] have reported that once particle size is reduced to less than 60 mesh, further reduction in size does not affect the counting efficiency, though many others[8,9] feel that sieving of the $Ba^{14}CO_3$ prior to incorporation into gel as unnecessary.

The relatively large counting efficiency obtained for direct particulate suspensions could be used advantageously. Hayes *et al.*[3] reported the first evidence that liquid scintillation method could be successfully used for the measurement of materials in suspension, rather than in solution. In their studies, $Ba^{14}CO_3$ was finely ground, moistening with ethanol before incorporation into the toluene scintillator which seems to be unnecessary from the present studies. Larger particle sizes, even after settling to the bottom, could be counted without much loss in efficiency. Table I shows the efficiency ratios of direct counting of varied particle sizes of $Ba^{14}CO_3$. There is very little difference in the efficiency for large particle sizes ($675 < \times < 1000\ \mu$ dia.) and the finest particle sizes employed ($74 < \times < 105\ \mu$ dia.) since the scintillator solution diffuses through the pores of the particles of $Ba^{14}CO_3$ and ^{14}C betas do not see much of the absorbing material before they produce photons. The good results of the method make it attractive at least for the measurement of high count rates.

The aqueous counting of $Na_2{}^{14}CO_3$ with sufficient inactive carrier has some disadvantages. When $8 \times 10^{-3}\ \mu ci$ containing 1 mg Na_2CO_3 was counted in 10 ml scintillator, the activity gradually decreased and attained a stable value very slowly. Heterogeneous system of counting, *viz.*, suspension with gel, is well suited even in such cases where the specific activity of the sample is less, since large amounts can be used without giving rise to this phosphorescence effects.

Quenching due to aluminum stearate in solution and in the gel form for tritiated toluene is given in Fig. 3. The nett efficiency obtained for ^{3}H, when 1·5 g aluminum stearate gel is formed in 10 ml toluene base scintillator, is 0·67 times that of solution counting. The self-absorption loss of tritium soft betas (18 kev) is so predominant even with lysine powder that it could be counted by suspension technique only with very small efficiency, 1% of the solution counting. Since the solution counting efficiency for tritium itself is 25%, the absolute counting efficiency for suspension technique will be of the order of 0·25%. Wang[10] also obtained counting efficiency for tritium as low as 1% when a paper strip containing absorbed tritium activity was dipped directly in a scintillator solution. The gel counting efficiency is low for tritium, but the linearity with respect to the total activity is well maintained. Thus gel counting for tritium is found to be not as attractive as that for carbon-14.

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